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Title: Uranium and Plutonium Isotopic Ratio Methods using FRAM

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Uranium and Plutonium Isotopic Ratio Methods using FRAM

Fixed energy

Response function

Analysis with

Multiple efficiencies

Pete Karpus

FRAM Isotopic Analysis Software

- FRAM is a powerful isotopic analysis code that has the same underlying theory as MGA.
- Fixed-energy Response-function Analysis with Multiple efficiencies
- Self-calibration using several gamma-ray peaks
- Analyze gamma ray data from 30keV to >1MeV of HPGe planar, HPGe Coaxial, or CdTe detector
- The key difference between the MGA family of codes and FRAM is the use of **user-editable analysis parameters** in the latter.

What Can FRAM Do?

- Control data acquisition
- Automatically analyze newly acquired data
- **Analyze previously acquired data**
- Provide information on the quality of the analysis
- Facilitate analysis in unusual situations
 - Non-standard energy calibrations
 - Gamma rays from non-SNM isotopes
 - Poor spectra (within limits)

Controlling Data Acquisition

- FRAM is not MCA software; it requires additional software to control basic detector settings
 - High Voltage
 - Pulse Shaping
- FRAM can be used to set acquisition times in real or live time
- FRAM shows you the spectrum as it is acquired if FRAM is controlling acquisition
 - Count time
 - Number of counts in the (moveable) cursor channel

Isotopic Ratio Principle

$$\frac{N^i}{N^k} = \frac{C(E_j^i)}{C(E_1^k)} \times \frac{T_{1/2}^i}{T_{1/2}^k} \times \frac{BR_1^k}{BR_j^i} \times \frac{RE(E_1)}{RE(E_j)}$$

N^i = Number of atoms of isotope i

$C(E_j^i)$ = Photopeak area of gamma ray j with energy E_j emitted from isotope i

$T_{1/2}^i$ = Half-life of isotope i

BR_j^i = Branching ratio (gamma rays/disintegration) of gamma ray j from isotope i

$RE(E_j)$ = Relative detection efficiency of gamma ray with energy E_j . Includes detector efficiency, measurement geometry, sample self-absorption, and attenuation in materials between the sample and detector.

Relative Efficiency

- General Expression for Peak Area Count Rate

$$\mathbf{C} = \frac{\mathbf{N} \times \ln 2 \times \mathbf{BR} \times \mathbf{RE}}{\mathbf{T}_{1/2}}$$

- Rearrange

$$\frac{\mathbf{C}}{\mathbf{BR}} = \left\{ \frac{\mathbf{N} \times \ln 2}{\mathbf{T}_{1/2}} \right\} \times \mathbf{RE}$$

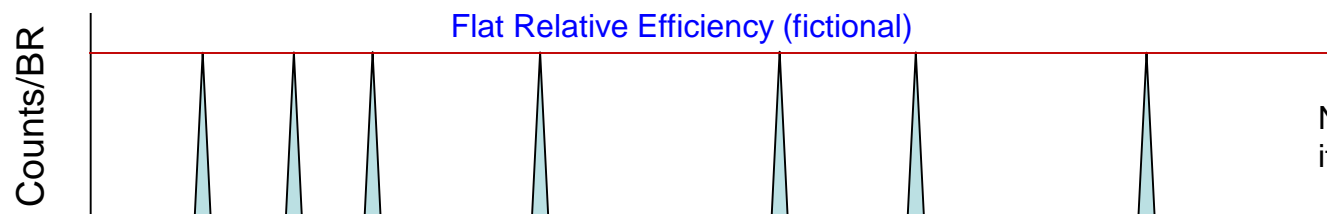
- Relative efficiency is proportional to count rate and branching ratio

$$\mathbf{RE} \propto \frac{\mathbf{C}}{\mathbf{BR}}$$

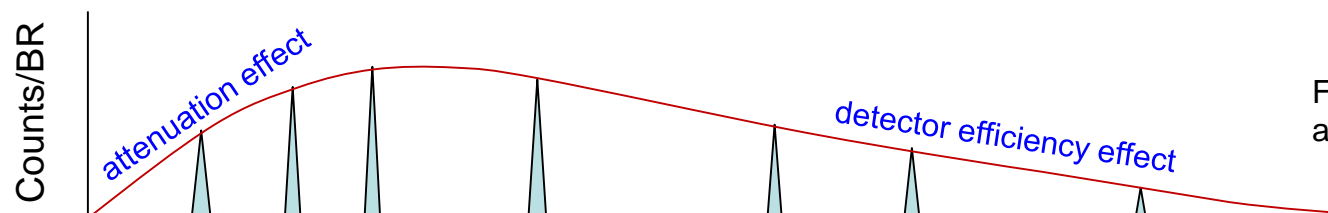
Relative Efficiency (Fictional Spectrum)



Detector has same efficiency at all energies (fictional).
No external or self attenuation.
Peak height only depends on branching ratios.



Now normalize each peak area by its branching ratio.

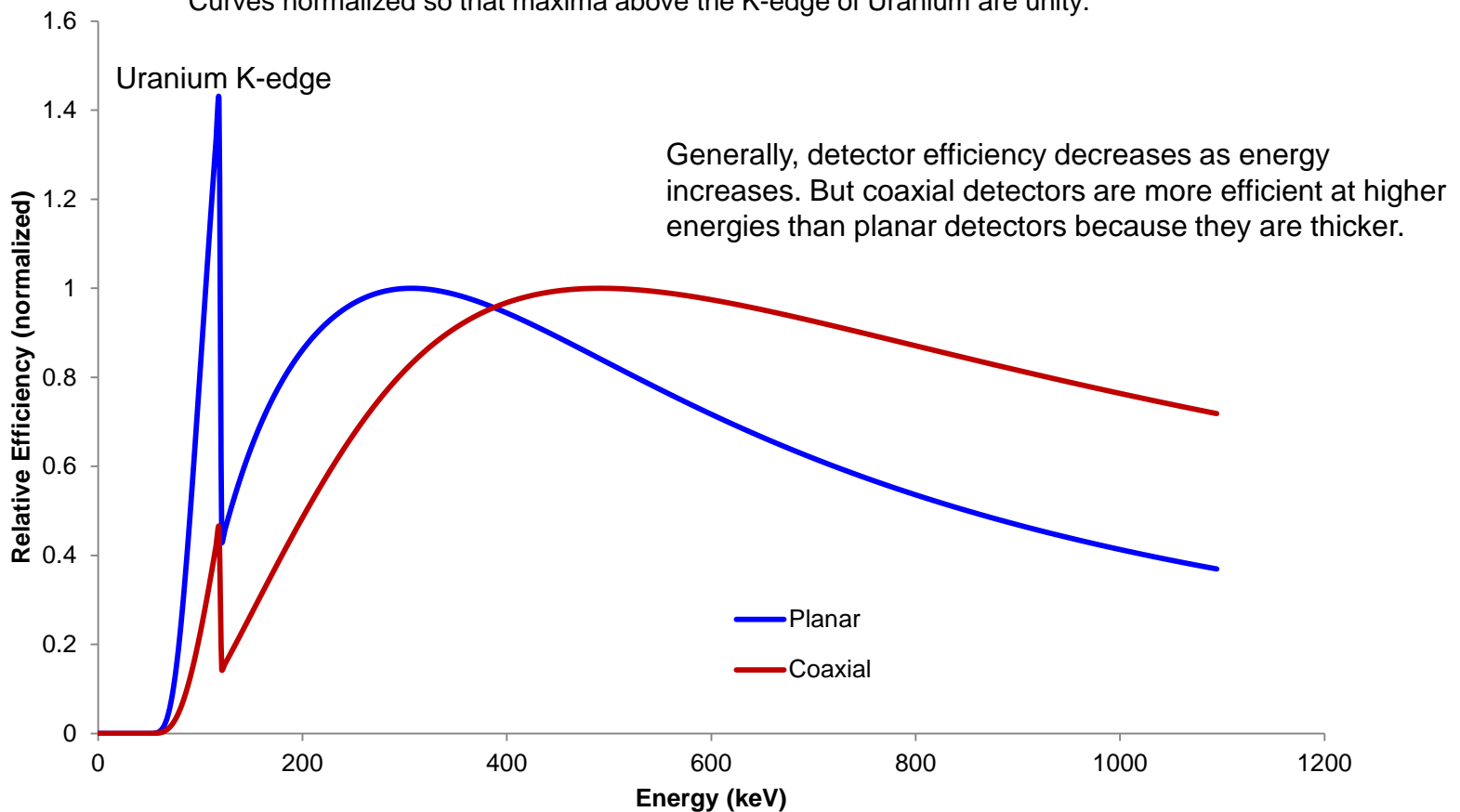


Finally, consider the real effects of attenuation and detector efficiency.

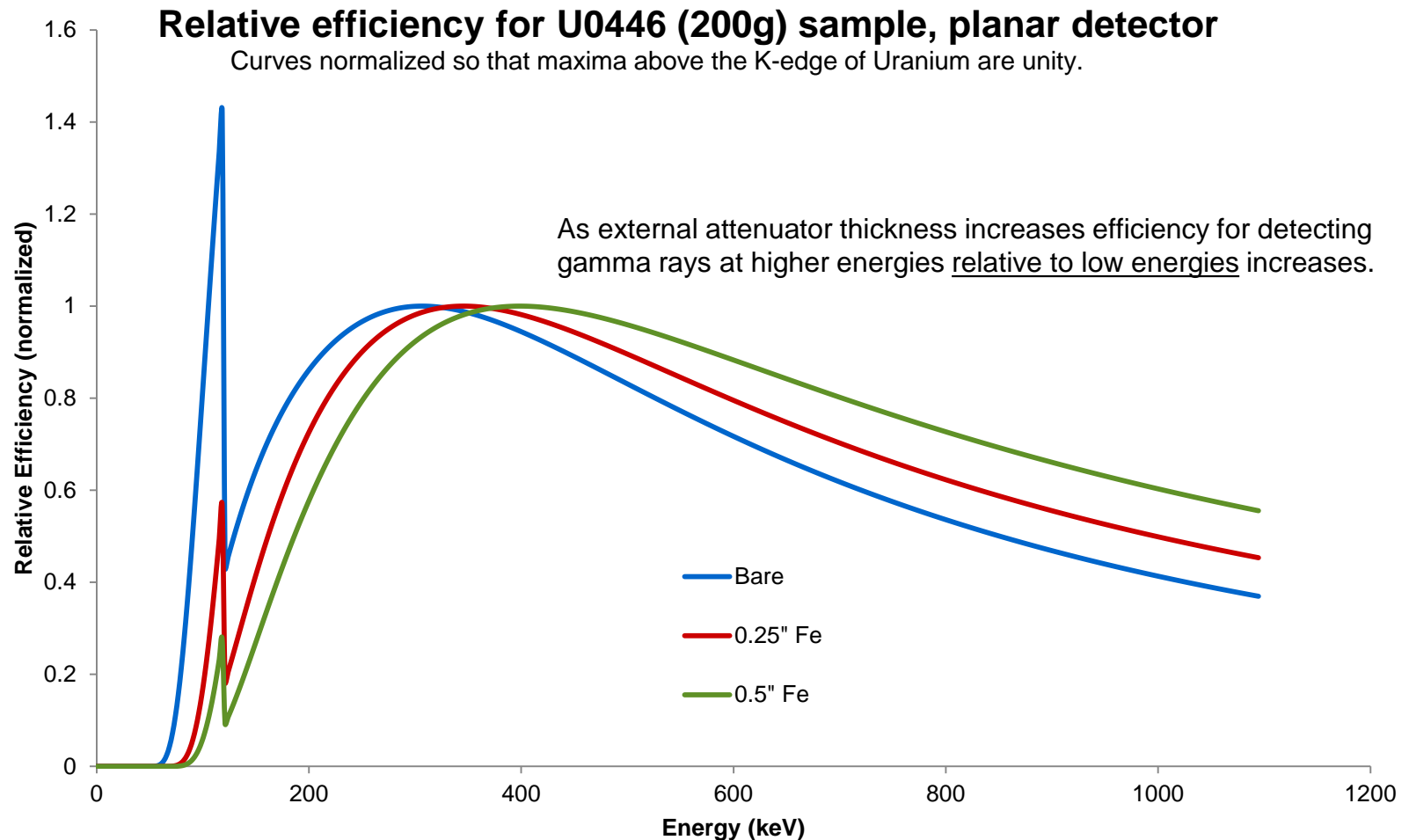
Relative Efficiency Depends on Detector Efficiency

Relative efficiency for Sample U0446 (200g), no absorber

Curves normalized so that maxima above the K-edge of Uranium are unity.



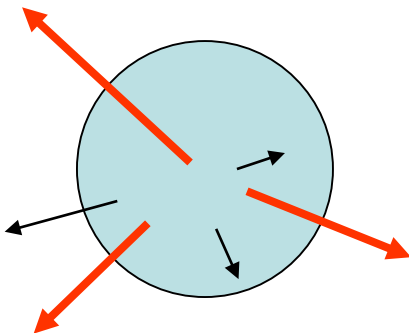
Relative Efficiency Depends on External Attenuation



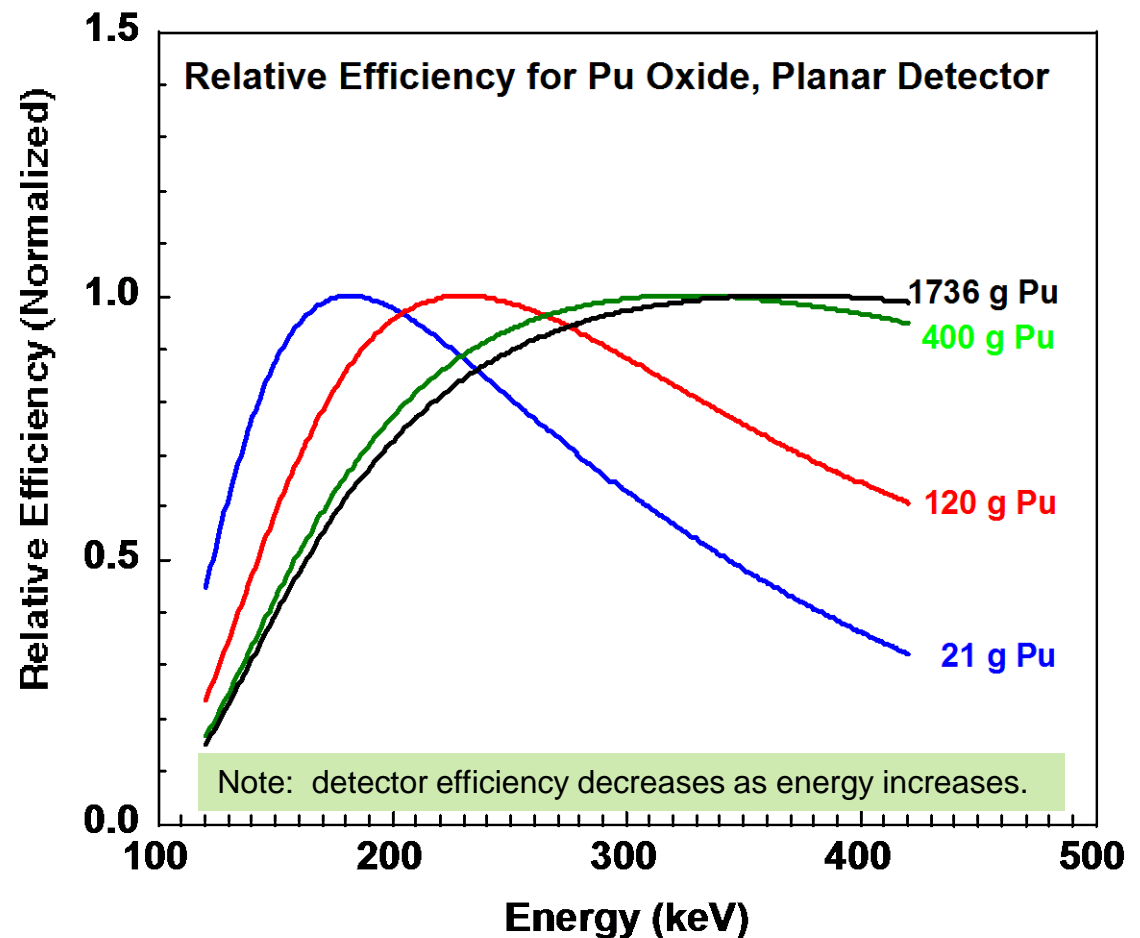
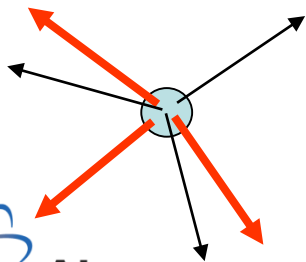
Relative Efficiency Depends on Self Attenuation

→ = Low E γ Rays
→ = High E γ Rays

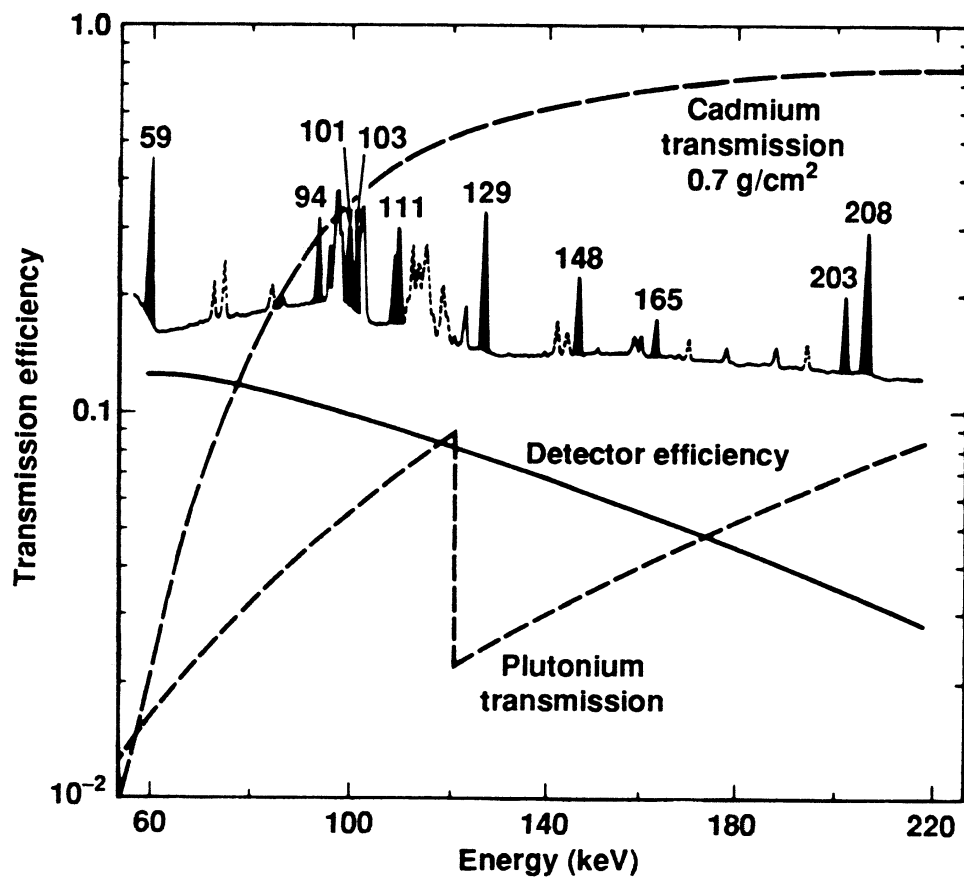
Large Mass Item



Small Mass Item



Relative Efficiency Curve Components



Independence of Analysis

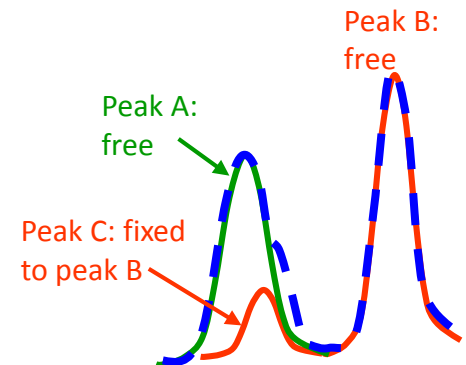
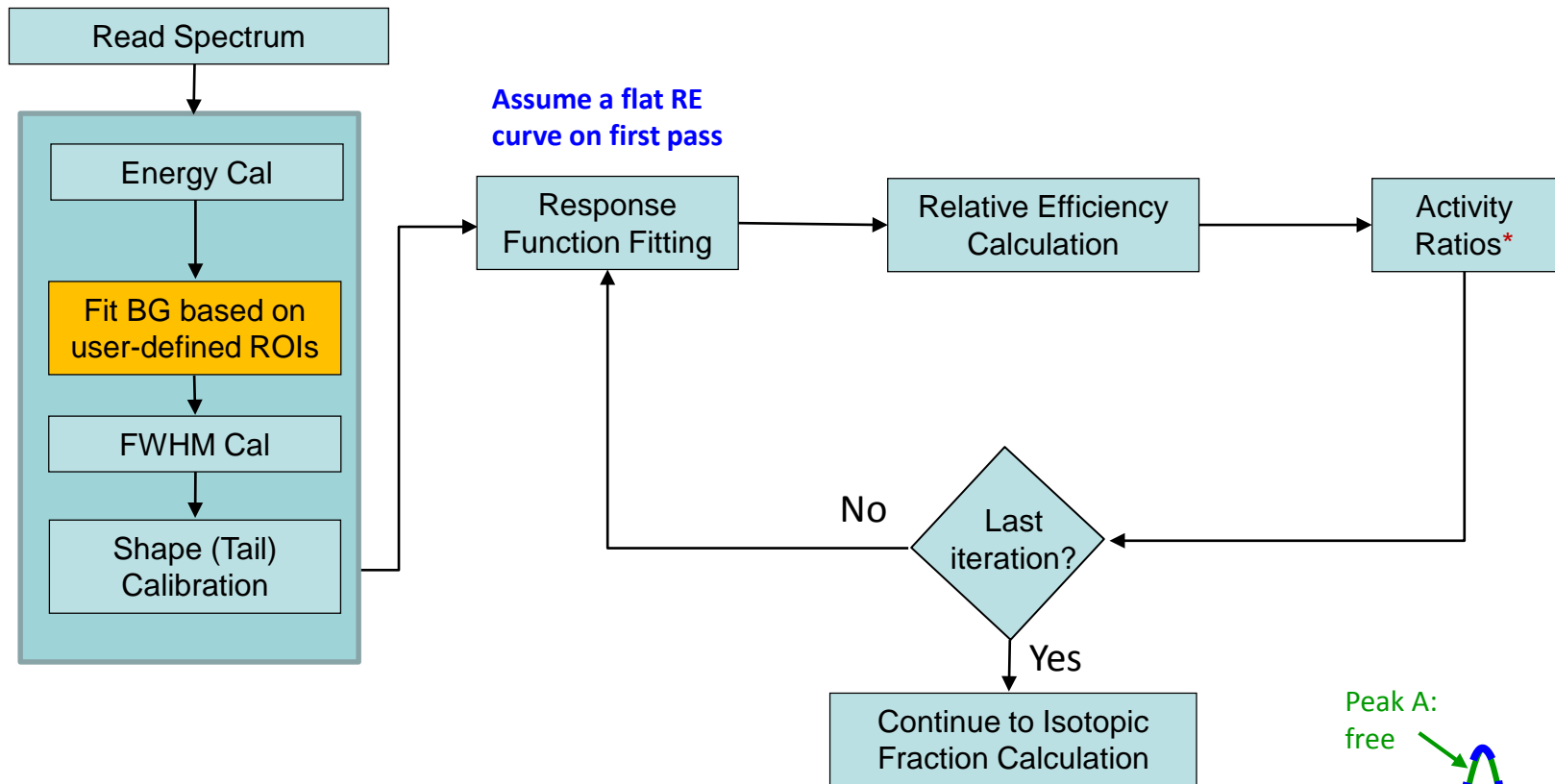
- Using **relative** efficiency and isotope **ratios** makes analysis independent of:
 - **Item Characteristics**
 - Size
 - Shape
 - Physical and Chemical Composition
 - Packaging
 - Filtering
 - **Data Acquisition Limitations**
 - Pulse Pile-up
 - Deadtime

NO CALIBRATION IS NEEDED!

FRAM Methodology

- **Internal Calibration**
 - Energy
 - FWHM
 - Peak Shape (Tailing Parameters)
- **Analysis**
 - Background continuum
 - Peak areas (Response Functions)
 - Relative Efficiency
 - Isotopic Ratio Calculation
 - Isotopic Mass Fraction Calculation

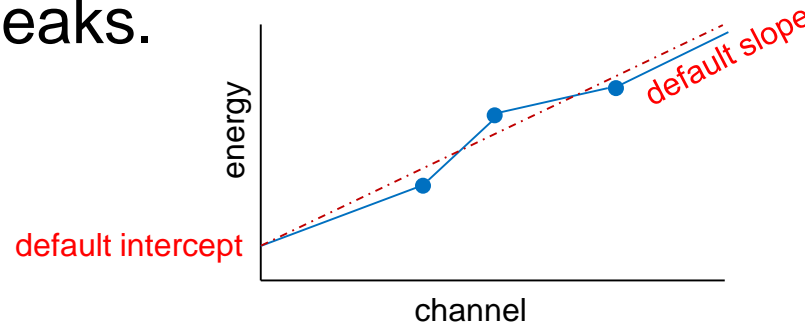
FRAM Top-Level Flow to Calculation of Activity Ratios



*Note: activity ratio calculations are needed in the iterations on the RE curve as peak stripping is often applied using multiple nuclides, which make up the data for the RE curve.

Internal Calibration

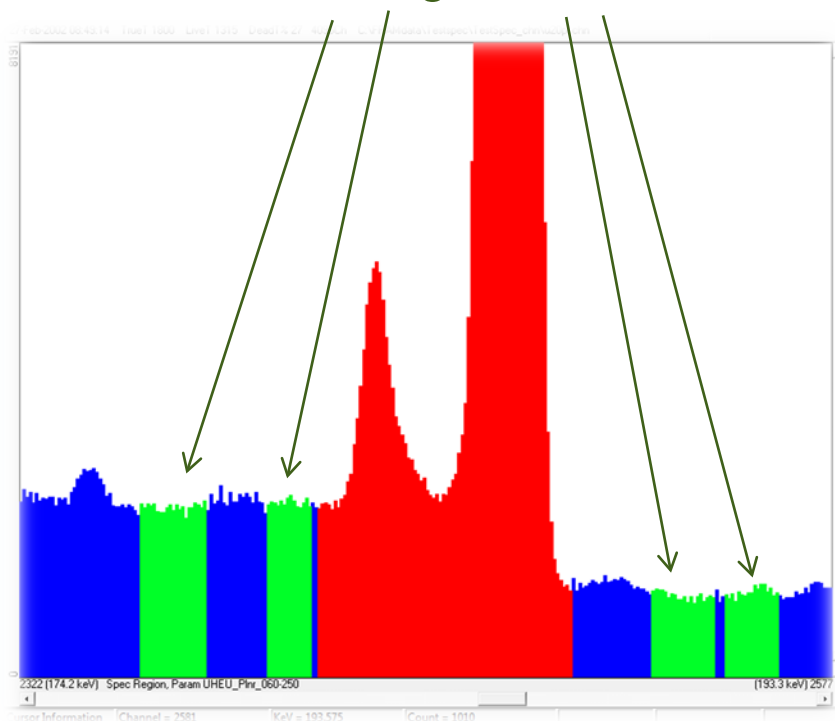
- **Energy** -- Piecewise linear calibration between designated peaks.



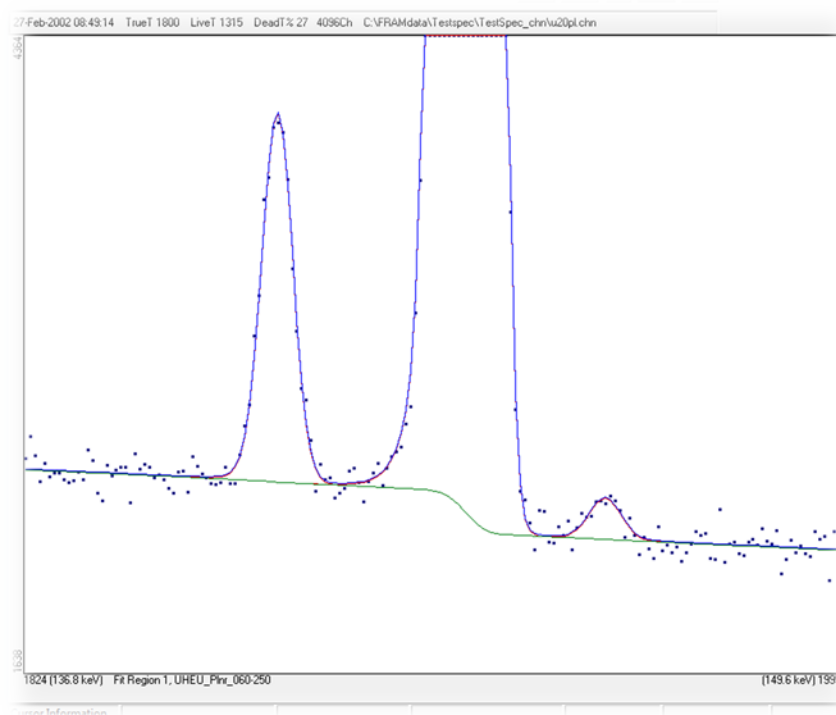
- **FWHM**² = $A_1 + A_2 * E + A_3 * E^{-1}$ fit to FWHM of designated peaks
- **Shape** -- Gaussian with exponential tail -- tail parameters found from residuals on designated peaks after subtracting Gaussian component.

Background Continuum

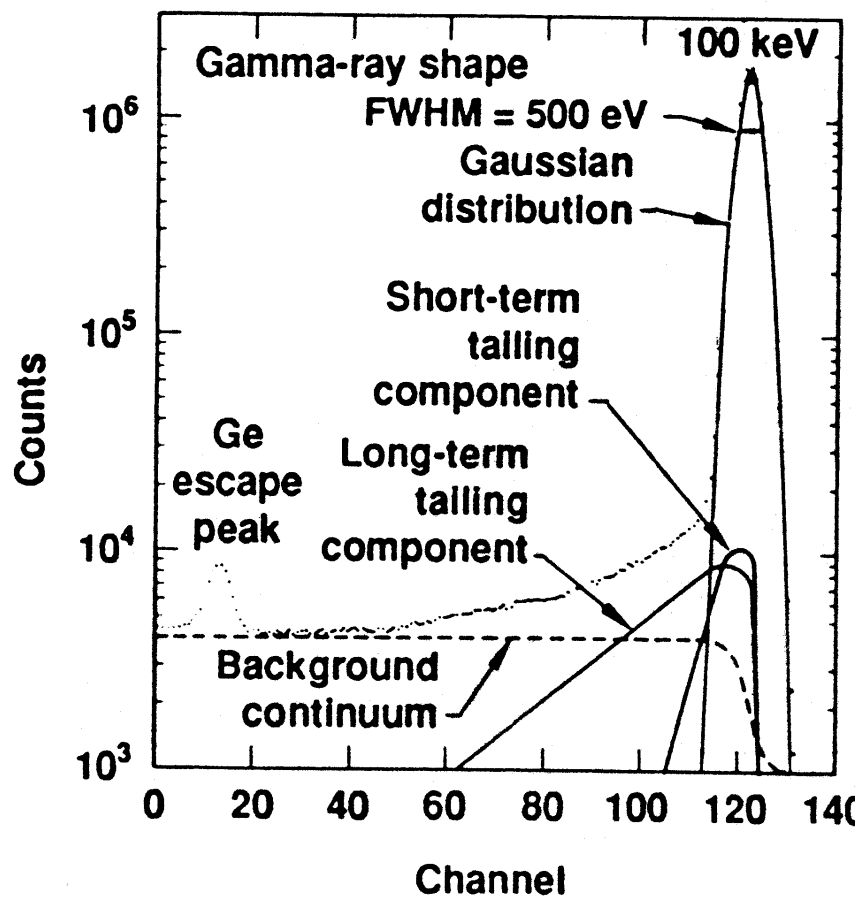
Continuum Regions of Interest



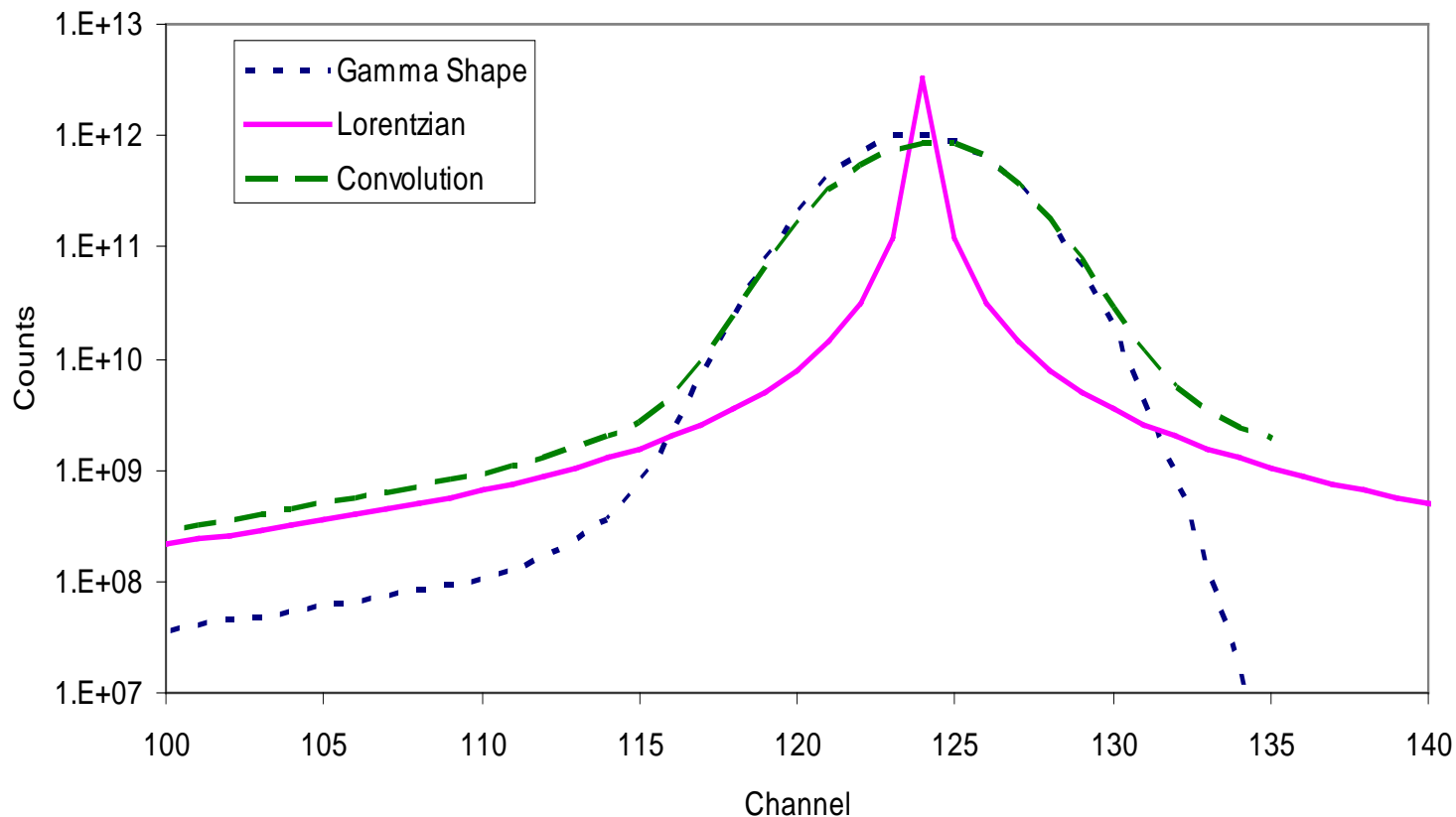
Calculated Step Function



Gamma-Ray Peak Fitting



X-Ray Peak Shape Convolution (Voigt)



Relative Efficiency Models

- Empirical or Physical
- The empirical model works well with good statistics within a range of continuity.
- The physical model is based on our understanding of radiation shielding physics
- It requires more knowledge
 - Density of the nuclear material
 - Major shielding materials and thicknesses
- The physical model can cross efficiency discontinuities (such as the K edge of Pu)

Empirical Relative Efficiency Curve

$$Y = C_1 + C_2 E^{-2} + C_3 (\ln E) + C_4 (\ln E)^2 + C_5 (\ln E)^3 + C_i + C_j E^{-1}$$

Y = log of ratio of (peak area/branching ratio)

E = energy

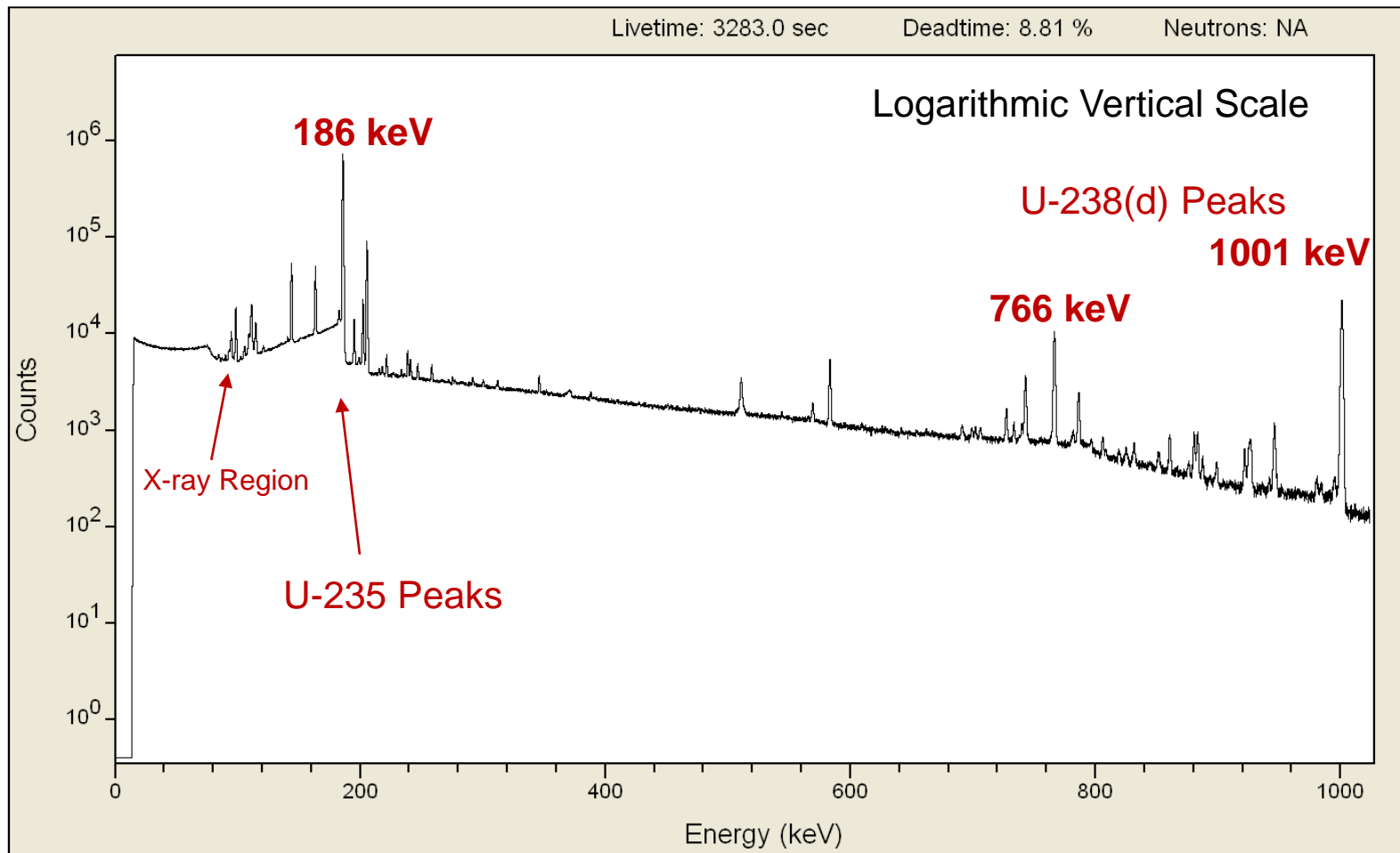
C_i = normalization for isotopes beyond the first one

C_j = normalization for different relative efficiency curves

Physical Efficiency Model

$$RE_i = \underbrace{\frac{1 - e^{-\mu_{Pu} \cdot x_{Pu}}}{\mu_{Pu} \cdot x_{Pu}}}_{\text{Self Attenuation}} \cdot \underbrace{\left(\prod_k e^{-\mu_k \cdot x_k} \right)}_{\text{External Attenuation (where } k = \text{Pb, Cd, Fe)}} \cdot \underbrace{I_i}_{\text{Intensity of } i^{\text{th}} \text{ Isotope}} \cdot \underbrace{e^{\beta_i / E}}_{\text{Heterogeneity Factor (associates additional efficiency functions beyond the first one)}} \cdot \underbrace{DetEff \cdot E^b c^{1/E}}_{\text{Hörl Function (adjustment for variation in detector efficiency and matrix)}}$$

Uranium Gamma-Ray Spectrum (1-MeV Range)



Uranium Isotopic Composition from Peak Ratios

- From the spectrum, measure the ratios:
 - $^{238}\text{f}/^{235}\text{f} \leftrightarrow \text{Isotopic Ratio}(^{238}\text{U}/^{235}\text{U})$
 - $^{234}\text{f}/^{235}\text{f} \leftrightarrow \text{Isotopic Ratio}(^{234}\text{U}/^{235}\text{U})$
- Then calculate the ^{235}U fraction using the constraint:

$$1 = ^{238}\text{f} + ^{235}\text{f} + ^{234}\text{f} + ^{236}\text{f}$$

i.e.

$$^{235}\text{f} = (1 + ^{238}\text{f}/^{235}\text{f} + ^{234}\text{f}/^{235}\text{f} + (^{236}\text{f}/^{235}\text{f}))^{-1}$$

- Use the ^{235}U fraction to solve for ^{238}f and ^{234}f .

^{236}U Estimate by Correlation

- Determine relative content of ^{236}U from a correlation function based on the fractions of the other uranium isotopes.
- Example form of a correlation function to determine the fraction of ^{236}U from the fractions of ^{235}U and ^{238}U :

$$^{236}\text{f} = A \cdot (^{235}\text{fB} + ^{238}\text{fC})$$

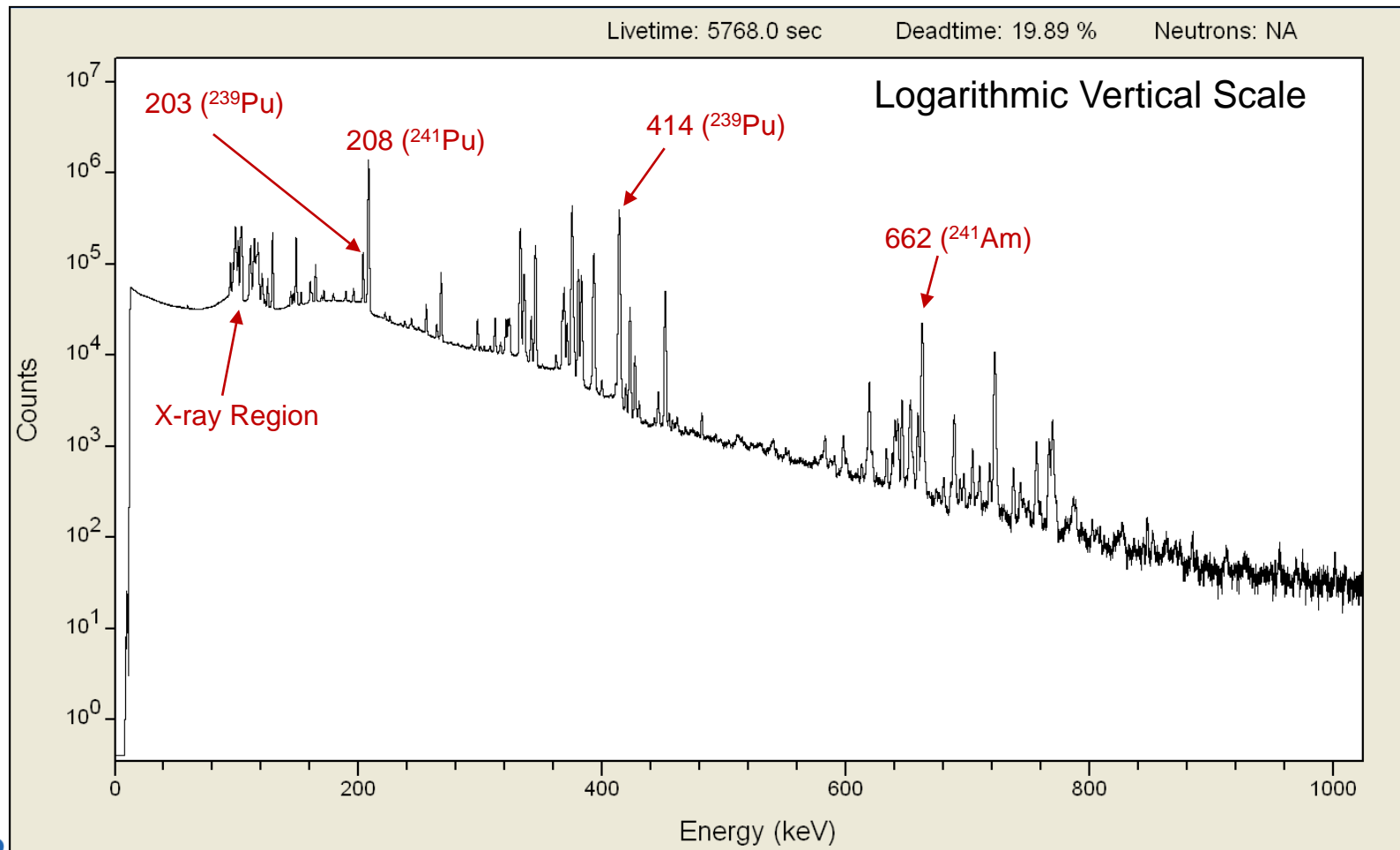
Ref. Los Alamos National Laboratory Document LA-UR-11-03005

- Once we have the fraction of ^{236}U , we can put it back into:

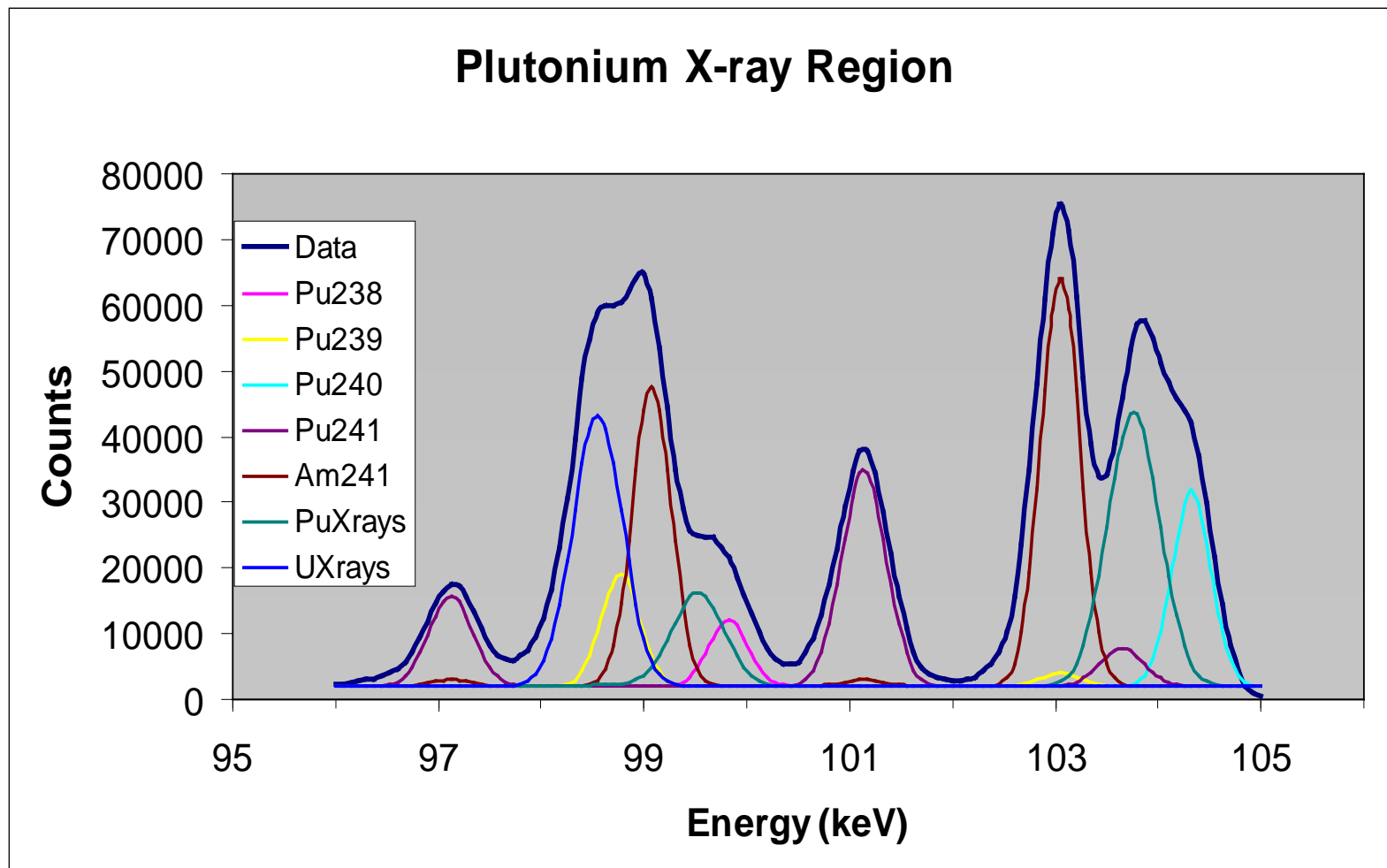
$$^{238}\text{f} + ^{235}\text{f} + ^{234}\text{f} + ^{236}\text{f} \geq 1$$

...and then renormalize ^{238}f , ^{235}f , and ^{234}f by this new sum.

Plutonium Gamma-Ray Spectrum (1-MeV Range)



Plutonium Response Function Analysis



Plutonium Isotopic Fractions from Isotopic Ratios

- Measure isotopic ratios : $^{238}\text{f}/^{239}\text{f}$
 $^{240}\text{f}/^{239}\text{f}$
 $^{241}\text{f}/^{239}\text{f}$
($^{242}\text{f}/^{239}\text{f}$)

- Sum of all fractions must equal 1:

$$1 = ^{238}\text{f} + ^{239}\text{f} + ^{240}\text{f} + ^{241}\text{f} + (^{242}\text{f})$$

- Divide by ^{239}f and then solve for ^{239}f :

$$^{239}\text{f} = [^{238}\text{f}/^{239}\text{f} + 1 + ^{240}\text{f}/^{239}\text{f} + ^{241}\text{f}/^{239}\text{f} + (^{242}\text{f}/^{239}\text{f})]^{-1}$$



^{242}Pu Estimate by Correlation

- Determine relative content of ^{242}Pu from a correlation function based on the fractions of the other plutonium isotopes:

$$^{242}\text{Pu} = 53[^{240}\text{Pu}] \times [^{241}\text{Pu}] / [^{239}\text{Pu}]^2$$

Gunnink, Nucl. Matl. Mgmt. 9, (2), 83-93 (1980)

Typical form of correlation functions used by MGA (& *FRAM*):

$$^{242}\text{Pu} = A \times [(^{238}\text{Pu})^B \times (^{239}\text{Pu})^C \times (^{240}\text{Pu})^D \times (^{241}\text{Pu} + ^{241}\text{Am})^E]$$

Requirements for Isotopic Analysis

- **Isotopic Homogeneity**
 - All plutonium (or uranium) must have the same isotopic composition even if it is not physically or chemically alike
- **Pu and Am must have same spatial distribution**



Isotopic Homogeneity

- The isotopic composition must be uniform throughout the item, even if it is chemically or physically non-uniform.
 - One exception is that FRAM can analyze physically uniform, but isotopically heterogeneous, matrices of plutonium and ^{241}Am .

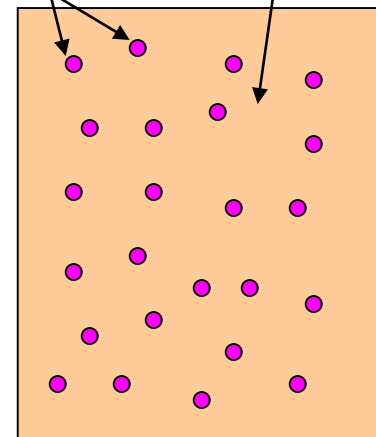
Isotopic Homogeneity:

Metal 80% Pu239 15% Pu240
Powder 80% Pu239 15% Pu240

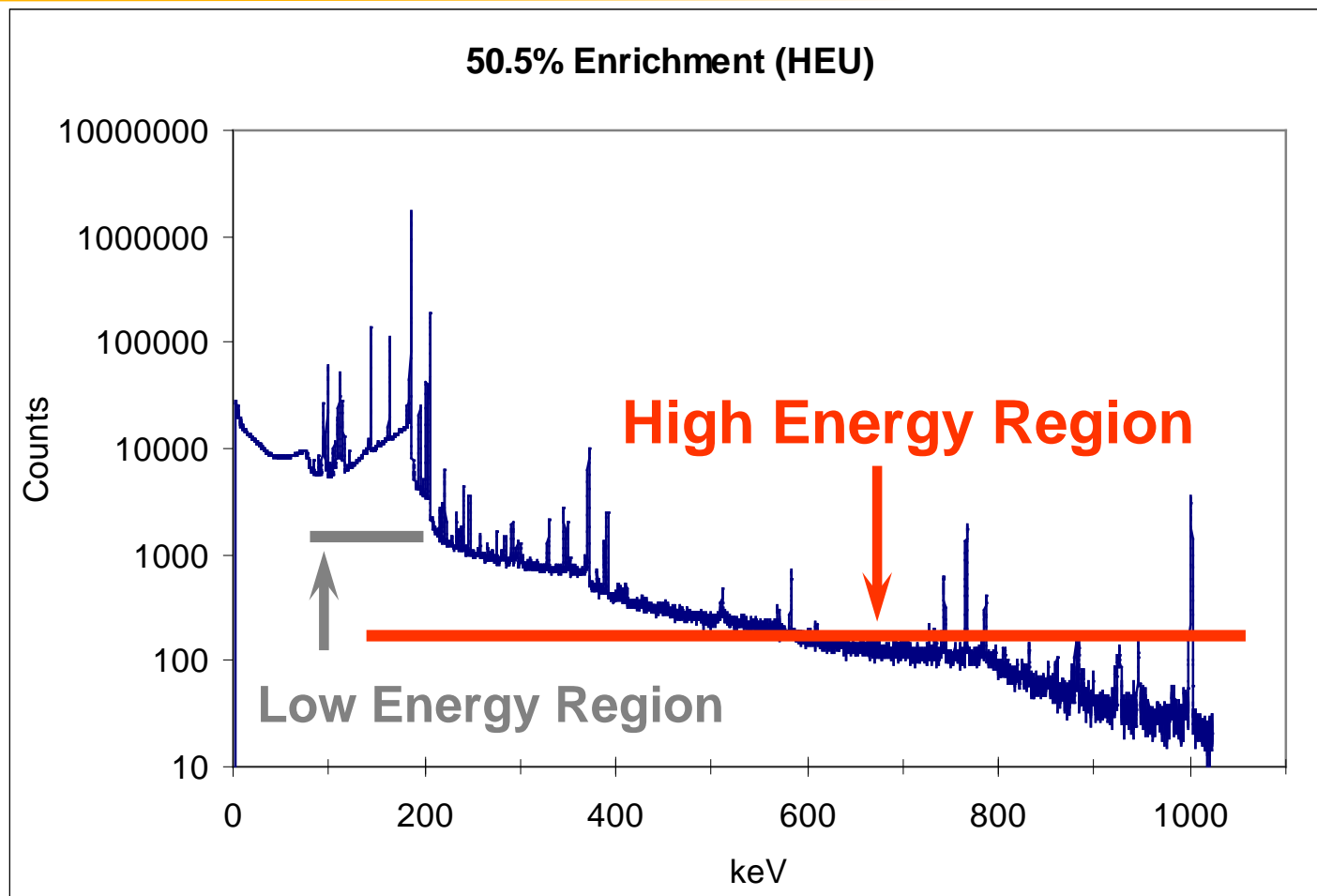
Isotopic Heterogeneity:

Powder 80% Pu239 15% Pu240
Powder 50% Pu239 45% Pu240

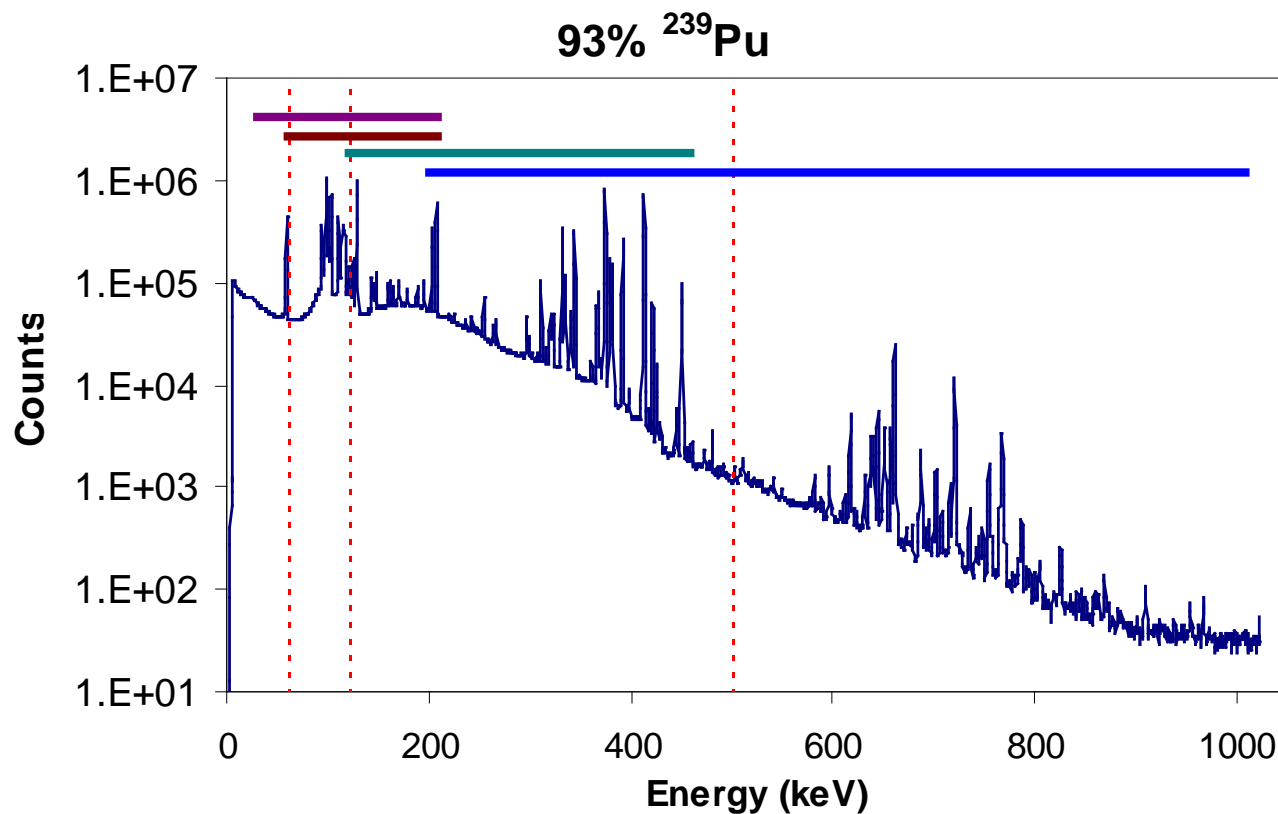
The Exception: Pu-metal in Am241 salt



Uranium Analysis Regions of FRAM



Plutonium Analysis Regions of FRAM



The four overlapping analytical regions that FRAM normally uses for the analysis are shown as four thick horizontal bars above the spectrum.

FRAM Analyze Window

The screenshot shows the 'Analyze' dialog box with the following fields and options:

- File:** C:\FRAMdata\Testspec\TestSpec_chn\u20pl.chn (Annotated: Spectrum File)
- Format:** Ortec 'chn' (dropdown)
- Parameter:** UHEU_Plnr_060-250 (dropdown) (Annotated: Parameter set)
- Gain/Offset:** 0.075 KeV/Ch + 0 KeV (Annotated: Default gain and offset)
- Number of Spectra:** 1
- Comment:** (empty text box)
- Efficiency Options:**
 - ☒ Pu242/U236 by correlation
 - ☐ Pu242/U236 by operator entry
 - ☐ Pu242/U236 by measurement
 - ☐ Empirical Efficiency
 - ☒ Physical Efficiency(Annotated: Efficiency Curve Option)
- % by weight:** 0
- Buttons:** Save Results, Print Results, Efficiency defaults
- Result File:** (empty text box)
- Analysis Options:**
 - ☐ Auto analysis
 - ☒ Uranium Analysis of Fresh Uranium (Annotated: For uranium not in secular equilibrium)
 - ☐ Previous Calorimetric measurement
 - ☐ Previous Neutron measurement
- Buttons:** OK, Cancel

FRAM (Short) Results Window

Raw data info

Parameter set

Errors/Warnings

Isotopic Fractions

Relative masses of other nuclides to U or Pu

FRAM 5.2 --- UserID: --- Power User

File Measure User Parameter Param Edit Help

☐ SpecOnly ☐ Region ☐ Ecal ☐ Fcal ☐ Scal ☐ fill ☐ log

☐ Efficiency ☐ Fits ☒ Short Result ☐ Med Result ☐ Long Result ☐ line ☐ lin

Parameter: UHEU_Plnr_060-250 0.075 KeV/Ch + 0 KeV

27-Feb-2002 08:49:14 TrueT 1800 LiveT 1315 DeadT% 27 4096Ch C:\FRAMdata\Testspec\TestSpec_chn\u20pl.chn

PC FRAM (5.2) Isotopic Analysis 10-Jan-2018 10:44:48
(Fixed energy Response function Analysis with Multiple efficiencies)
Operator ID:

spectrum source: C:\FRAMdata\Testspec\TestSpec_chn\u20pl.chn
spectrum date: 27-Feb-2002 08:49:14
live time: 1315 s
true time: 1800 s
num channels: 4096

parameter set: UHEU_Plnr_060-250 (2013.07.25 10:17)
U Only, Enrichment >=10%, 0.075 keV/ch, Planar Detector
Physical Efficiency, Gain 0.075 keV/ch, Offset 0 keV
comment:

diagnostics passed.

	(OpEntry)			
	U234	U235	U236	U238
mass%	0.1461	19.9859	0.0000	79.8680
sigma	0.0016	0.1090	0.0000	0.1098
%RSD	1.09%	0.55%	>99.99%	0.14%
%TotPwr	92.81	4.25	0.00	2.83

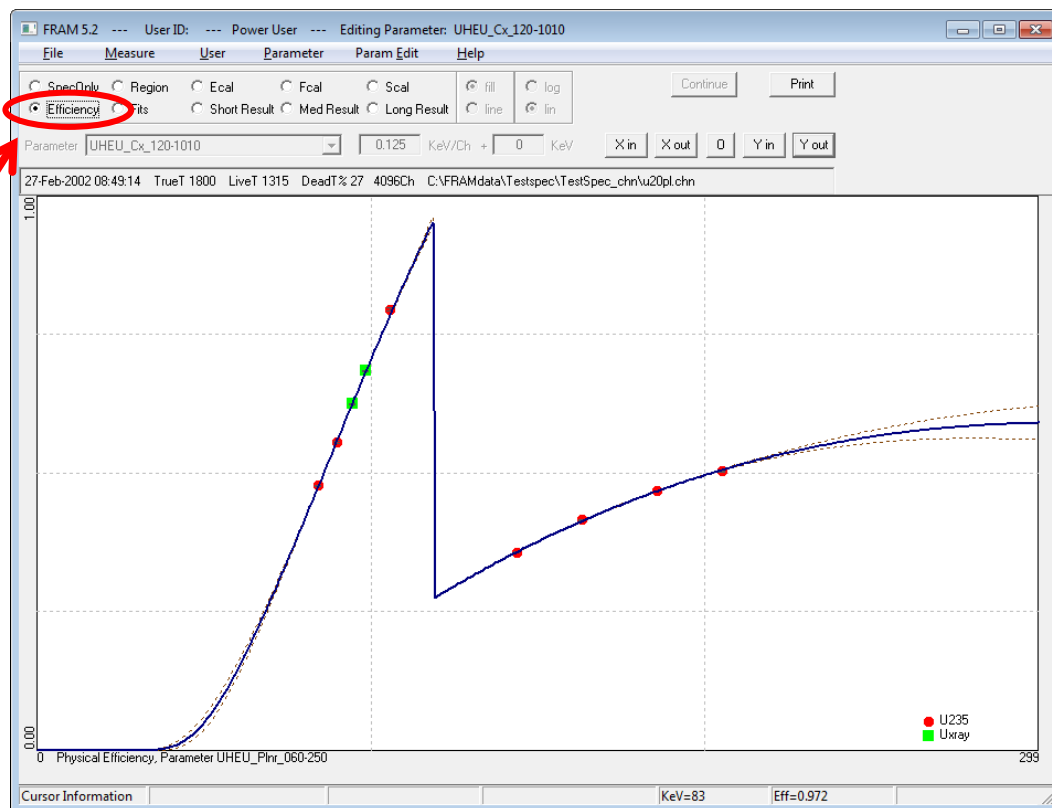
Specific Power (W/gU): (0.2821 +/- 0.0029)e-006 (1.01%)

Relative mass (Th228 / U): 1.713e-012 (1.90%)
Relative mass (Uxray / U): 3.470e-001 (0.40%)

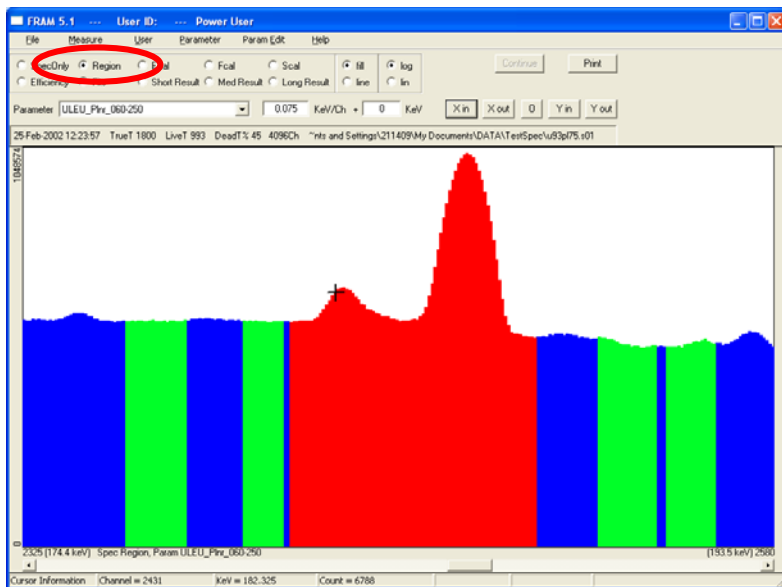
Note: For Plutonium you will see "time since ²⁴¹Am separation" in this location

FRAM Diagnostics

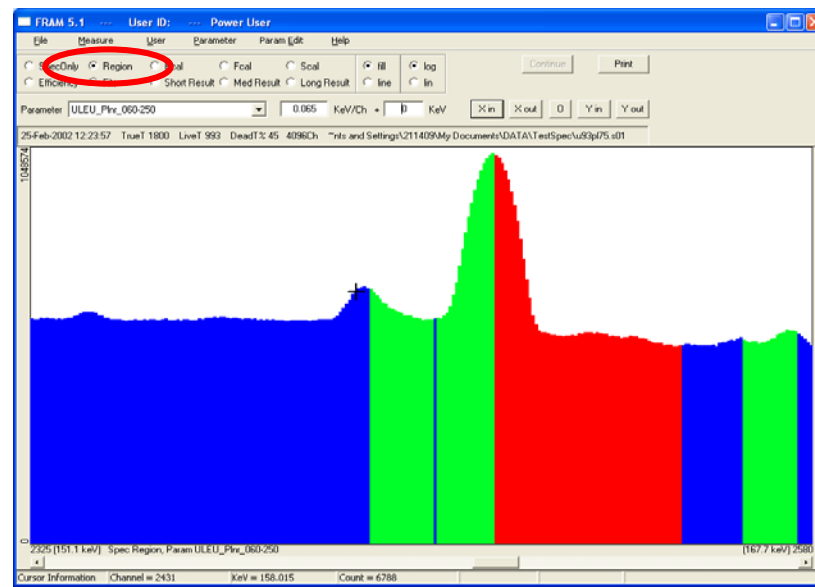
- FRAM provides diagnostic tools for the user to check the quality of the analysis
- **Diagnostic Tools**
 - Spectrum
 - Fits
 - Relative Efficiency
 - Results



Region: Checking the GAIN

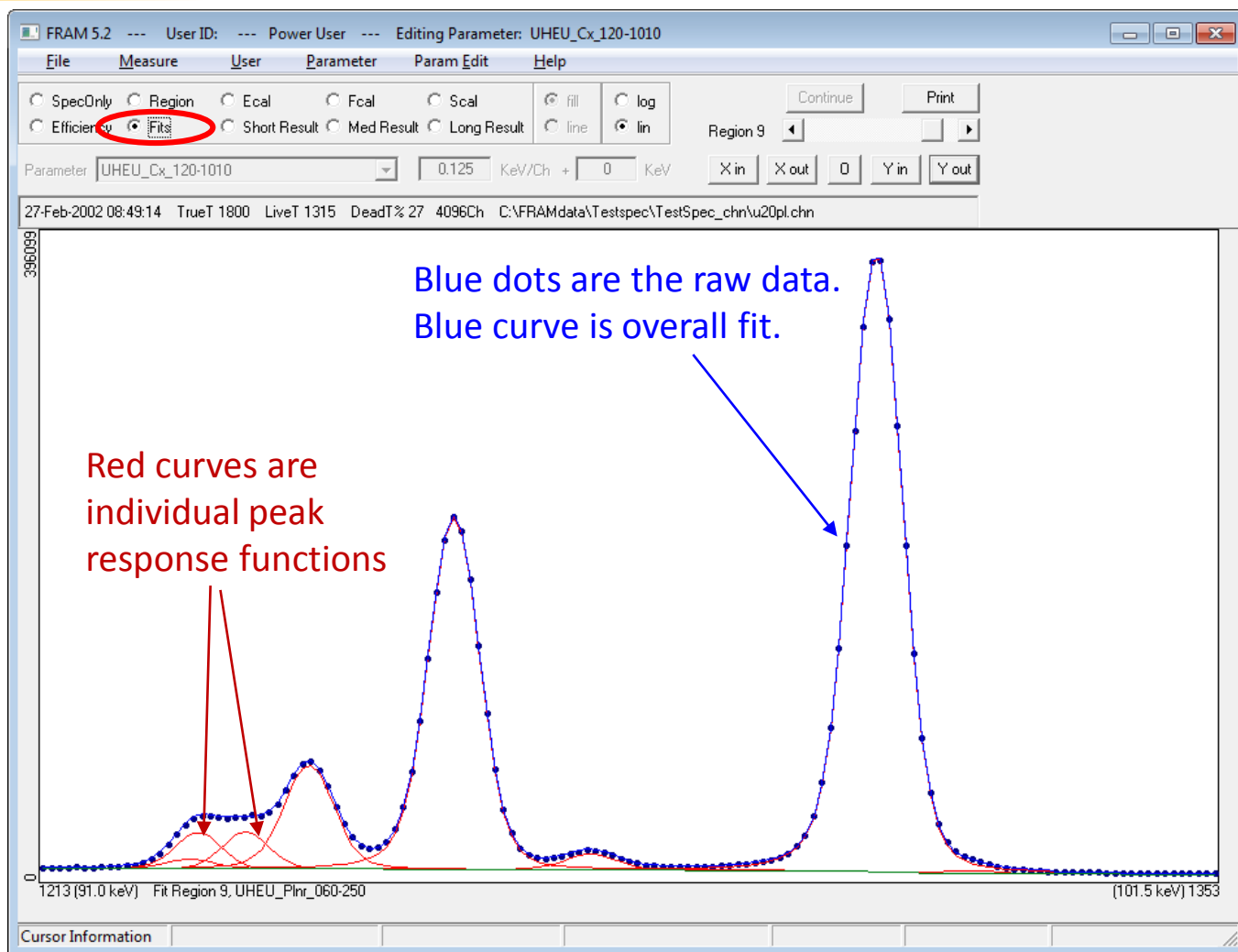


ROIs aligned with peaks
Default ECAL is OK



ROIs NOT aligned with peaks
Check default ECAL

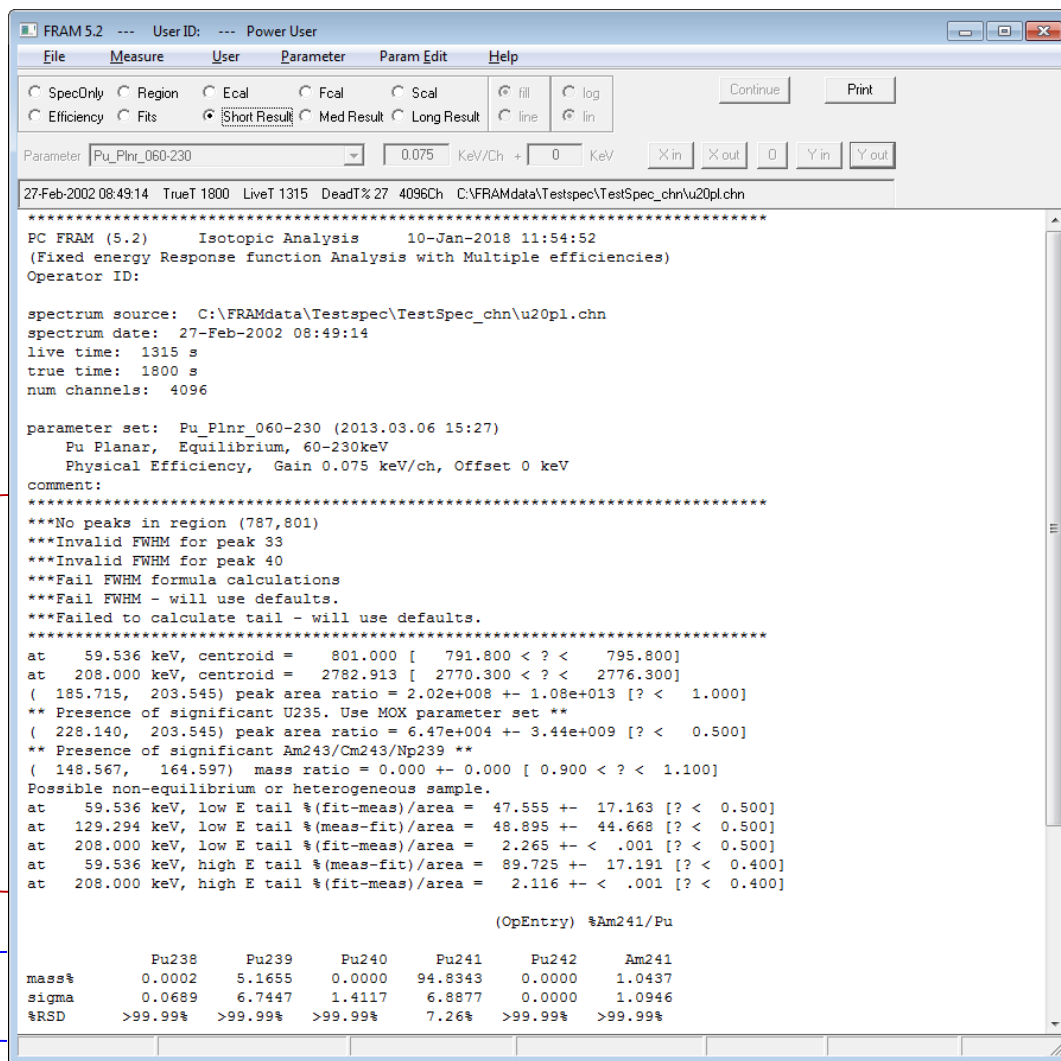
Fits: Sum of All Peaks in Region



FRAM Results (Example: Major Errors)

Now we have tons of error messages and warnings

Isotopic Fractions are nonsensical



The screenshot shows the FRAM 5.2 software window. The title bar indicates 'User ID: Power User'. The menu bar includes File, Measure, User, Parameter, Param Edit, and Help. The toolbar contains buttons for SpecOnly, Region, Ecal, Fcal, Scal, fill, log, Efficiency, Fits, Short Result (selected), Med Result, Long Result, line, lin, Continue, and Print. The Parameter field is set to 'Pu_Plnr_060:230'. The report text is as follows:

```
27-Feb-2002 08:49:14 TrueT 1800 LiveT 1315 DeadT% 27 4096Ch C:\FRAMdata\Testspec\TestSpec_chn\u20pl.chn
*****
PC FRAM (5.2) Isotopic Analysis 10-Jan-2018 11:54:52
(Fixed energy Response function Analysis with Multiple efficiencies)
Operator ID:

spectrum source: C:\FRAMdata\Testspec\TestSpec_chn\u20pl.chn
spectrum date: 27-Feb-2002 08:49:14
live time: 1315 s
true time: 1800 s
num channels: 4096

parameter set: Pu_Plnr_060-230 (2013.03.06 15:27)
Pu Planar, Equilibrium, 60-230keV
Physical Efficiency, Gain 0.075 keV/ch, Offset 0 keV
comment:
*****
***No peaks in region (787,801)
***Invalid FWHM for peak 33
***Invalid FWHM for peak 40
***Fail FWHM formula calculations
***Fail FWHM - will use defaults.
***Failed to calculate tail - will use defaults.
*****
at 59.536 keV, centroid = 801.000 [ 791.800 < ? < 795.800]
at 208.000 keV, centroid = 2782.913 [ 2770.300 < ? < 2776.300]
( 185.715, 203.545) peak area ratio = 2.02e+008 +- 1.08e+013 [? < 1.000]
** Presence of significant U235. Use MOX parameter set **
( 228.140, 203.545) peak area ratio = 6.47e+004 +- 3.44e+009 [? < 0.500]
** Presence of significant Am243/Cm243/Np239 **
( 148.567, 164.597) mass ratio = 0.000 +- 0.000 [ 0.900 < ? < 1.100]
Possible non-equilibrium or heterogeneous sample.
at 59.536 keV, low E tail %(fit-meas)/area = 47.555 +- 17.163 [? < 0.500]
at 129.294 keV, low E tail %(meas-fit)/area = 48.895 +- 44.668 [? < 0.500]
at 208.000 keV, low E tail %(fit-meas)/area = 2.265 +- < .001 [? < 0.500]
at 59.536 keV, high E tail %(meas-fit)/area = 89.725 +- 17.191 [? < 0.400]
at 208.000 keV, high E tail %(fit-meas)/area = 2.116 +- < .001 [? < 0.400]

(OpEntry) %Am241/Pu

mass% Pu238 Pu239 Pu240 Pu241 Pu242 Am241
sigma 0.0002 5.1655 0.0000 94.8343 0.0000 1.0437
%RSD 0.0689 6.7447 1.4117 6.8877 0.0000 1.0946
%RSD >99.99% >99.99% >99.99% 7.26% >99.99% >99.99%
```


FRAM Analyze Window for Detective Data

Set Parameter set to one designed for a coaxial detector

File: C:\Users\211409\Desktop\BB TESTSPECTRA\Detective_U20.CHN Browse...

Format: Ortec 'chn'

Parameter: UHEU_Cx_120-1010 0.366 KeV/Ch + 0 KeV

Number of Spectra: 1

Comment:

☒ Pu242/U236 by correlation ☐ Empirical Efficiency
☐ Pu242/U236 by operator entry % by weight: 0 ☒ Physical Efficiency
☐ Pu242/U236 by measurement Efficiency defaults

☐ Save Results ☐ Print Results

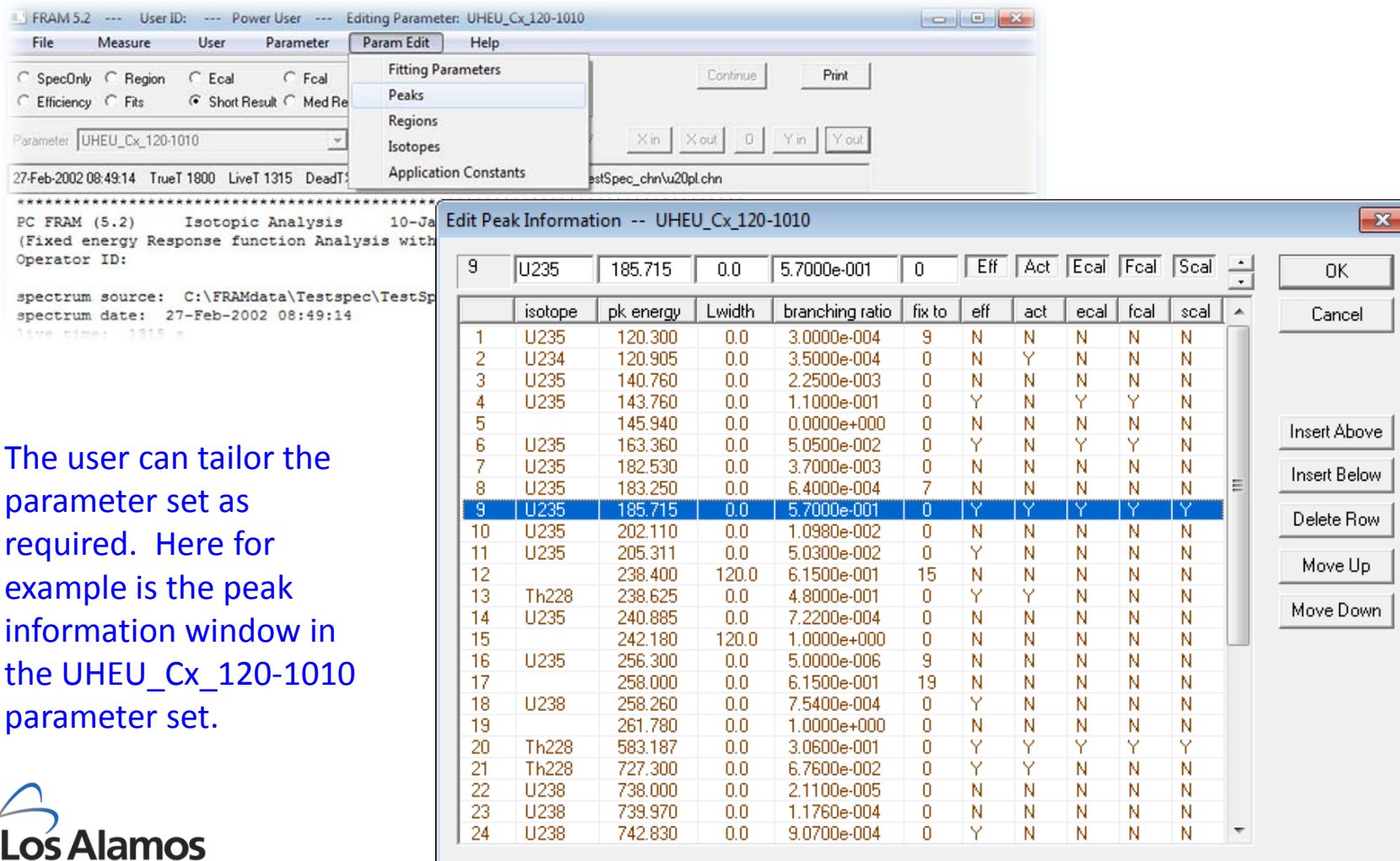
Result File: Browse...

☐ Auto analysis
☐ Uranium Analysis of Fresh Uranium
☐ Previous Calorimetric measurement ☐ Previous Neutron measurement

OK Cancel

Set default gain to 0.366 keV/ch

Editing Parameter Sets



FRAM 5.2 --- User ID: --- Power User --- Editing Parameter: UHEU_Cx_120-1010

File Measure User Parameter Param Edit Help

☐ SpecOnly
 ☐ Region
 ☐ Ecal
 ☐ Fcal
 ☐ Efficiency
 ☐ Fits
 ☒ Short Result
 ☐ Med Re

Parameter: UHEU_Cx_120-1010

27-Feb-2002 08:49:14 TrueT 1800 LiveT 1315 DeadT

PC FRAM (5.2) Isotopic Analysis 10-Ja

(Fixed Energy Response function Analysis with Operator ID:

spectrum source: C:\FRAMdata\Testspec\TestSp

spectrum date: 27-Feb-2002 08:49:14

live time: 1315 s

Edit Peak Information -- UHEU_Cx_120-1010

	isotope	pk energy	Lwidth	branching ratio	fix to	eff	act	ecal	fcal	scal
9	U235	185.715	0.0	5.7000e-001	0	Eff	Act	Ecal	Fcal	Scal
1	U235	120.300	0.0	3.0000e-004	9	N	N	N	N	N
2	U234	120.905	0.0	3.5000e-004	0	N	Y	N	N	N
3	U235	140.760	0.0	2.2500e-003	0	N	N	N	N	N
4	U235	143.760	0.0	1.1000e-001	0	Y	N	Y	Y	N
5	U235	145.940	0.0	0.0000e+000	0	N	N	N	N	N
6	U235	163.360	0.0	5.0500e-002	0	Y	N	Y	Y	N
7	U235	182.530	0.0	3.7000e-003	0	N	N	N	N	N
8	U235	183.250	0.0	6.4000e-004	7	N	N	N	N	N
9	U235	185.715	0.0	5.7000e-001	0	Y	Y	Y	Y	Y
10	U235	202.110	0.0	1.0980e-002	0	N	N	N	N	N
11	U235	205.311	0.0	5.0300e-002	0	Y	N	N	N	N
12	U235	238.400	120.0	6.1500e-001	15	N	N	N	N	N
13	Th228	238.625	0.0	4.8000e-001	0	Y	Y	N	N	N
14	U235	240.885	0.0	7.2200e-004	0	N	N	N	N	N
15	U235	242.180	120.0	1.0000e+000	0	N	N	N	N	N
16	U235	256.300	0.0	5.0000e-006	9	N	N	N	N	N
17	U235	258.000	0.0	6.1500e-001	19	N	N	N	N	N
18	U238	258.260	0.0	7.5400e-004	0	Y	N	N	N	N
19	U238	261.780	0.0	1.0000e+000	0	N	N	N	N	N
20	Th228	583.187	0.0	3.0600e-001	0	Y	Y	Y	Y	Y
21	Th228	727.300	0.0	6.7600e-002	0	Y	Y	N	N	N
22	U238	738.000	0.0	2.1100e-005	0	N	N	N	N	N
23	U238	739.970	0.0	1.1760e-004	0	N	N	N	N	N
24	U238	742.830	0.0	9.0700e-004	0	Y	N	N	N	N

OK Cancel

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The user can tailor the parameter set as required. Here for example is the peak information window in the UHEU_Cx_120-1010 parameter set.

Examples

- Uranium Examples:
 - Clean analysis of good spectrum
 - Analysis with incorrect gain
 - Enrichment Study (0.3, 20, 91%)
 - Shielded Item
- Plutonium Examples
 - Clean analysis of good spectrum
 - Analysis in different energy regions
 - Shielded Item
 - Correlation Function Limitations
 - Interferences / Additional Nuclides